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01/15/99
ASS't Commissioner of Patents
Washington, D.C. 20231

ATT: APPLICATION BRANCH

Re: U.S. Application of Giuseppe GUARINO and Ermanno FILIPPI for
"METHOD FOR IN-SITU MODERNIZATION OF A HETEROGENEOUS SYNTHESIS
REACTOR"

Our Ref: Q-52856

01/15/99
JC518 U.S. PRO
09/23/1991
01/15/99

Sir:

Attached hereto is the application identified above including the claim to priority, executed Declaration, SES Declaration, Preliminary Amendment, Assignment, Information Disclosure Statement and drawings.

The Government filing fee is calculated as follows:

Total claims	10 - 20 = 00	x \$18(09) =
Independent Claims	02 - 03 = 00	x \$78(39) =
Base Fee (\$760.00/\$380.00)		380.00
Assignment Recordal Fee (\$40.00)		40.00
Multiple Dependent Claim Fee (\$260./\$130.)		
Foreign Language Filing Fee (\$130.00)		
TOTAL FILING FEE		420.00

Attached is a check in the amount of \$420.00 for the initial filing of the application and recordal of the Assignment. The Patent Office is authorized to charge or credit any difference or overpayment to Deposit Account No. 19-4880. The Commissioner is hereby authorized to charge any fees under 37 C.F.R. §§ 1.16 and 1.17 which may be required during the entire pendency of the application to Deposit Account No. 19-4880.

Priority is claimed for this application from European applications 98200114.1 filed January 16, 1998. The priority document will be submitted in due course.

Respectfully submitted,

Robert V. Sloan

Reg. No. 22,775

RVS:jlg

Attachments:

PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of Giuseppe GUARINO et al.

Filed: January 15, 1999

For: METHOD FOR IN-SITU MODERNIZATION OF A HETEROGENEOUS SYNTHESIS REACTOR

Ass't Commissioner of Patents
Washington, D.C. 20231

ATT: APPLICATION BRANCH

Sir:

PRELIMINARY AMENDMENT

Prior to the examination of the above identified application and to the calculation of the fees, please amend this application as follows:

IN THE SPECIFICATION:

Page 4, change "proving" to -- providing --.

IN THE CLAIMS:

Claim 1, line 15, change "proving" to -- providing --.

REMARKS

The above changes have been amended to correct a typographical error in the specification and in claim 1. An early and favorable action on the merits is respectfully requested.

Respectfully submitted,



Robert V. Sloan
Reg. No. 22,775

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Date: January 15, 1999

Att'y Docket No: Q-52856

Title: Method for in-situ modernization of a heterogeneous synthesis reactor"

DESCRIPTION

Application field

5 The present invention relates to a method for in-situ modernization of a heterogeneous synthesis reactor, in particular for the exothermic synthesis such as ammonia or methanol synthesis and the conversion of carbon monoxide, including at least a catalytic bed of the radial or axial-
10 radial type, provided with opposite cylindrical perforated walls for the inlet and outlet of gases.

In the description given below and in the following claims, the term: "in-situ modernization", is understood to mean the on-site modification of a pre-existing reactor in order
15 to improve its performances and obtain e.g. a production capacity and/or a conversion yield comparable to those of a newly-built reactor.

In the terminology of the field, this type of modernization is also termed retrofitting or revamping.

20 As is known, in the field of heterogeneous synthesis reactions in general, the need is increasingly felt of adapting the pre-existing synthesis reactors to the catalysts of new conception with always increasing reaction activity, in order to achieve an improvement in the
25 conversion yield and a reduction in energy consumption, while lowering investment costs.

In fact, the continuous progress in the realisation of high activity catalysts has caused - the production capacity of the reactor being the same - the catalyst mass to be loaded
30 in the respective bed to be markedly lower than the maximum filling volume for which the bed was designed, allowing therefore savings in the cost of said catalyst.

Prior art

In pre-existing reactors provided with catalytic beds of the axial type, the adaptation of the reactor to the new high reaction activity catalysts does not cause special problems, as the catalytic bed(s) may be loaded with a more or less high quantity of catalyst without involving substantial modifications in the running of the same - in particular from the fluid dynamics point of view - except for a different pressure drop than can be anyhow regulated by suitably modifying the operating conditions of the reactor.

In pre-existing reactors comprising catalytic beds of the radial or axial-radial type, the loading of a catalyst mass different from the design mass involves on the contrary severe drawbacks in the running of the catalytic bed(s).

A catalytic bed of the radial type only partly filled with catalyst, unavoidably presents rows of holes in the gas inlet and outlet walls which are uncovered in the upper part of the bed, with ensuing undesired bypass of the same by the reaction gases and a corresponding drastic reduction in the reactor conversion yield.

The same problem arises in a catalytic bed of the axial-radial type, wherein there also lacks the axial crossing of the catalyst by the reaction gases, which involves a further reduction in the conversion yield compared to an optimal loaded catalytic bed.

In particular, the presence of a reduced amount of catalyst in the axial-radial bed, besides uncovering a part of the holes of the perforated gas inlet and outlet walls, thwarts the function performed by the unperforated portion of the upper part of the gas outlet wall of axially routing the gases entering said bed.

Even though the so-called retrofitting of existing reactors

has become increasingly accepted, which technique aims at avoiding an expensive replacement of the latter while achieving at the same time the maximum conversion yield and the minimum energy consumption compatible with the 5 available reaction volume, no methods have been proposed as at today allowing the adaptation of existing reactors provided with catalytic beds of the radial or axial-radial type that can satisfy the above need.

At present, lacking valid technical solutions, the radial 10 or axial-radial catalytic beds of existing reactors are still loaded with conventional catalysts, to the detriment of the improvement in conversion yield and energy consumption, which would be instead achievable by utilising the new high reaction activity catalysts.

15 Otherwise, i.e. by using a high activity catalyst, it is always necessary to fill the available volume of the radial or axial-radial catalytic bed entirely, in order to prevent the aforementioned drawbacks, and accordingly there is obtained - besides an increase in the conversion yield - 20 also an increase in the production capacity of the existing reactor that is not always required or desired, as such increase may for instance involve a replacement of the apparatuses located downstream of the synthesis reactor, which would be otherwise undersized, with related high 25 investment and construction costs.

Besides, filling the whole volume available in the existing 30 catalytic beds which have been so designed as to be suitable to contain a conventional lower activity catalyst, requires an amount of high activity catalyst such as to render the investment cost prohibitive.

Because of these very drawbacks, the utilisation of high activity catalysts in pre-existing heterogeneous synthesis reactors comprising catalytic beds of a radial or axial-radial type, has not had till now a concrete application,

even though the need is increasingly felt in the field.

Summary of the invention

The problem underlying the present invention is that of providing a method for modernizing a heterogeneous synthesis reactor of the type comprising at least a radial or axial-radial bed such as to allow to use of new conception catalysts having always greater reaction activity, in order to achieve an improvement in conversion yield and a reduction in energy consumption, in a simple and reliable way and to low investment and operating costs.

Said problem is solved by a method of the type set forth above, characterised in that it comprises the following steps:

- providing an unperforated cylindrical wall coaxial to said gas outlet wall in said catalytic bed, said unperforated cylindrical wall extending from an upper end of said gas outlet wall for a portion of the same of a prefixed length, so as to define a free-space between the gas outlet wall and the unperforated wall, for the passage 15 of a part of the gas leaving said catalytic bed;
- proving means of closing said free-space between the unperforated wall and the gas outlet wall, in proximity of the upper end of the latter, preventing thereby a bypass of said catalytic bed or a recycling to the same of the gas 20 entering respectively leaving the reactor.

Advantageously, the method according to the present invention allows a partial loading of the pre-existing radial or axial-radial catalytic bed(s), allowing in this way an effective utilisation of the new high activity 30 catalysts, without affecting the running of the beds adversely, in particular keeping the fluid dynamics and pressure drop characteristics of the same unchanged.

In fact, thanks to presence of an unperforated wall of a prefixed length near the upper zone of the gas outlet wall and the simultaneous formation of a free-space between the unperforated wall and the gas outlet wall, it is
5 advantageously possible to reach a twofold aim, expounded hereunder.

On the one hand, the unperforated wall allows to route into the catalytic mass the gas flow entering the beds, preventing thereby the formation of undesired bypasses,
10 i.e. preventing gas from flowing directly through the holes of the gas outlet wall caused to be uncovered because of the only partial filling of the catalytic beds, without passing through the catalyst.

On the other hand, the presence of the free-space allows
15 the gas flow that has passed through the catalytic mass from escaping through all of the holes of the gas outlet wall, so as to keep the pressure drop through the catalytic bed(s) unchanged.

Particularly satisfactory results have been achieved by
20 providing an unperforated wall that extends for a portion comprised between 5% and 50% the length of the gas outlet wall, respectively defining a substantially annular free-space having a thickness comprised between 0,5 and 10 cm.

In this way it is possible to load even relatively low
25 amounts of high activity catalyst, without the risk of undesired catalytic bed bypasses by the synthesis gas, while keeping unchanged the fluid dynamics and pressure drop characteristics preceding the retrofitting.

With reference to the present invention, it is worth
30 stressing that being able to conceive an only partial loading of the catalyst in a catalytic bed of the radial or axial radial type - without affecting thereby the running of the same adversely - is in sharp contrast with the

constant teaching of the prior art according to which the use of radial or axial-radial catalytic beds inevitably involves a full filling of the same with catalyst, to prevent beds from being undesirably bypassed by reaction
5 gases.

In fact, because of the very intrinsic characteristics of such beds, loading only partly a radial or axial-radial catalytic bed was inconceivable according to the prior art.

Only after the research of the applicant, it has been
10 possible to solve the aforementioned technical problem, by proposing a modernization of the pre-existing catalytic radial or axial-radial beds that allows - contrary to the teaching of the prior art - a partial loading of the same.

15 The characteristics and advantages of the invention are set forth in the description of an example of implementation of a modernization method in accordance with the invention, given hereinbelow by way of non-limiting illustration with reference to the annexed drawing.

Brief description of the drawing

20 Figure 1 shows schematically a longitudinal section of an existing reactor for carrying out heterogeneous synthesis reactions, suitably modified according to the modernization method of the present invention.

Detailed description of a preferred embodiment

25 With reference to Figure 1, reference 1 indicates as a whole an heterogeneous synthesis reactor.

Reactors of this type are especially suitable for carrying out exothermic heterogeneous synthesis reactions at high pressure and temperature (20-300 bar, 180-550°C), for
30 instance for the production of ammonia or methanol or for the conversion of carbon monoxide into carbon dioxide.

Reactor 1 comprises a tubular envelope or shell 2, provided at the top with a nozzle 3 for the inlet of the reaction gases and at the bottom with a nozzle 4 for the outlet of the reaction products.

- 5 Shell 2 is also provided at the top with a nozzle 5 to allow the passage of a worker in the inside of reactor 1 to carry out the various operations of assembly and maintenance of the same. Nozzles of this type are generally known to those skilled in the art by the jargon name of
10 "manholes".

In the inside of shell 2 a catalytic bed 6 of the axial-radial type is obtained, defined sideways by the respective gas inlet and outlet cylindrical perforated walls 7 respectively 8, and underneath by the bottom of the shell
15 2.

- The catalytic bed 6 is not closed at the top to allow the same to be axially crossed by a portion of the reaction gases. To prevent undesired catalyst leakages, containment nets - generally known to those skilled in the art and
20 therefore not shown - may be installed in the catalytic bed 6.

In the example of Figure 1, the gas inlet wall 7 is located near shell 2, while the gas outlet wall 8 is located in the middle of reactor 1. Between shell 2 and gas inlet wall 6 a free-space 9 is obtained to allow a radial crossing of bed
25 6 by the reaction gases. Gas outlet wall 8 is also closed at the top by a gas-tight lid 10, of a known type.

- A chamber 11, extended coaxial to the catalytic bed 6, between wall 8 and lid 10, is lastly provided in reactor 1,
30 for routing the reaction products leaving said bed to nozzle 4, through which they are finally evacuated.

The broken line 12 shown in proximity of the upper end of the gas inlet wall 7 delimits the highest level that can be

reached by the catalyst in the inside of the catalytic bed 6, and defines, together with side walls 7 and 8 and the bottom of the shell 2, the reaction volume available in reactor 1.

- 5 Such volume has been calculated based on the reaction activity of the catalyst commercially available at the time of the design of reactor 1, to achieve a predetermined production capacity.

Therefore, before being modernized according to the present
10 invention, reactor 1 still had a catalytic bed 6 whose volume was entirely taken up by a conventional catalyst.

On the contrary, the broken line 13 indicates the level reached by the catalyst in reactor 1 advantageously modernized according to the present invention.

- 15 The catalyst in the inside of bed 6 is indicated - as a whole - by 14 and has a reaction activity such as to provide a production capacity of the reactor equivalent to the design capacity, but taking up a volume substantially smaller than the volume of the catalytic bed 6.
20 In other words, thanks to the greater reaction activity, the mass of catalyst 14 loaded in the reactor once the latter is modernized in accordance with the invention results to be - the production capacity being the same - much smaller than the catalyst mass employed before the
25 modernization, accordingly involving savings in the catalyst cost.

Arrows F of Figure 1 indicate the various routes followed by the gas through the catalytic bed 6.

- According to a first step of the modernization method of
30 the present invention, there is provided a substantially cylindrical unperforated wall 15 co-axial to the gas outlet wall 8 in the catalytic bed 6. The unperforated wall 15

protrudes from an upper end 8a of the gas outlet wall 8 for a pre-fixed portion of the same, so as to define an annular free-space 16 between the gas outlet wall 8 and the unperforated wall 15, for the passage of a part of the gas
5 leaving said catalytic bed 6, as indicated by arrows F of Figure 1.

In a further step of the present method, means are provided of closing the free-space 16 between the unperforated wall 15 and the gas outlet wall 8, in proximity of the upper end
10 8a of the latter, preventing thereby the bypass of the catalytic bed 6 or the recycling to the same of the gases entering respectively leaving the reactor.

Thanks to the steps of providing an unperforated wall near the upper end of the gas outlet wall, and of defining a
15 free-space between said walls for the passage of reacted gases, it is advantageously possible to load the catalytic bed with amounts of catalyst substantially lower than the design amounts, without affecting the running of the same adversely, in particular keeping its fluid dynamics and
20 pressure drop characteristics unchanged.

In fact, even though the level of catalyst 14 remains well beneath the upper end 8a of the gas outlet wall 8 (broken line 13), leaving therefore uncovered several holes of such wall, the unperforated wall 15 prevents gas reagents from
25 crossing the catalytic bed 6 without penetrating into the catalytic mass, and free-space 16 allows to utilise all the holes of wall 8 as outlets for the reaction products.

Should unperforated wall 15 be in touch directly with gas outlet wall 8 - without the formation of free-space 16 - a
30 catalytic bed would be obtained having the same fluid dynamics characteristics as the non-modernized bed, but, due to the reduction in the number of holes available for the outlet of the reaction products, the pressure drop would be increased.

In the example of Figure 1, unperforated wall 15 extends advantageously for a portion comprised between 20% and 40% the length of gas outlet wall 8. In practice, wall 15 extends preferably for such a length as to re-create in the
5 catalytic bed 6, only partly loaded with catalyst 14, a zone prevailingly axially crossed by the reaction gases.

Was the catalytic bed 6 of a merely radial type, wall 15 would arrive barely beyond the broken line 13 which defines the level reached by catalyst 14, so as to ensure a
10 substantially radial crossing of the catalytic bed.

Moreover, the free-space 16 is preferably so defined as to have a thickness comprised between 1 and 5 cm. In any case, the thickness of free-space 16 must be great enough to allow gas crossing without causing an additional pressure
15 drop.

Advantageously, free-space 16 is closed in proximity of the upper end 8a of the gas outlet wall, so as to prevent undesired bypasses of gas reagents entering the catalytic bed 8 or the recycling to the same of reaction products.

20 In order to simplify as much as possible the implementation of the present modernization method, the unperforated wall 15 is suitably supported by the gas outlet wall 8.

For instance, wall 15 may be removably fixed to wall 8 through special supporting means hooked to the latter in
25 proximity of its upper end 8a.

In particular, according to a preferred embodiment of the invention, shown in Figure 1, the unperforated wall 15 - whose diameter is greater than the gas outlet wall 8 - is advantageously supported by a horizontal gas-tight baffle
30 17 which protrudes above the upper end 8a of the gas outlet wall 8 and leans on the same.

Advantageously, wall 15 and baffle 8 form a kind of gas-

tight glass - for instance made from unperforated plate - which rests reversed on lid 10 of the gas outlet wall 8.

Upon conclusion of said steps, a reactor 1 is obtained which allows to perform heterogeneous synthesis reactions
5 with high conversion yields and at low energy consumption, in the following way.

Gas reagents, let into reactor 1 through nozzle 3, are fed to the catalytic bed 6 comprising a high activity catalyst 14.

10 Depending on the type of reaction, the temperature and pressure of the gas reagents fed to the catalytic bed 6 are regulated downstream of reactor 1.

The gas reagents cross the catalytic bed 6 with an axial-radial centripetal flow. Thanks to the presence of the
15 unperforated wall 15, it is possible to deviate the flow of gas reagents axially, preventing undesired bypasses of the catalytic bed 6.

The reaction products obtained in the catalytic bed 6 cross the gas outlet wall 8 and are afterwards collected in
20 chamber 11, to leave then finally reactor 1 through nozzle 4. A (minority) part of the reaction products advantageously flows along free-space 16 which allows in this way to exploit also the part of wall 8 circumscribed by wall 15 for the outlet of gases.

25 In so doing, it is possible - the production capacity of the pre-existing reactor being the same - to load only partly the catalytic bed 6 with a high reaction activity catalyst, obtaining savings in the cost of said catalyst, and keeping at the same time the fluid dynamics and
30 pressure drop characteristics of the catalytic bed unchanged.

If an increase in the production capacity of the pre-

existing reactor should be required - which would involve the necessity of fully exploiting the available volume of the catalytic bed 6, loading the same with a high-activity catalyst - it would suffice to take out from the reactor 5 the unperforated wall 15, and consequently also baffle 17 that supports the same, to bring back the catalytic bed 6 to its original configuration.

The present invention is advantageously applicable especially in the fields of heterogeneous synthesis 10 reactions where the technological progress has allowed to develop new catalysts having always increasing reaction activities.

A very interesting field is without any doubt the field of ammonia synthesis where, thanks to the present method, it 15 is now possible to effectively modernize the pre-existing reactors so as to use high activity catalysts such as graphite supported ruthenium-based catalysts.

Another particularly interesting field is the field of carbon monoxide conversion, where the pre-existing reactors 20 (for instance of the type shown in Figure 1) may be advantageously loaded with reduced volumes of high activity catalysts, such as for instance copper-comprising catalysts for high temperature conversion.

However, the modernization method according to the present 25 invention is not limited to the type of reactor described above with reference to Figure 1, but may be also applied to reactors comprising a plurality of radial or axial-radial beds supported for instance in the inside of an appropriate cartridge.

30 Besides, for the purposes of the implementation of the present method it does not matter at all whether the catalytic bed is crossed by the reaction gases with a centripetal or a centrifugal flow. In the latter instance,

the gas outlet wall 8 would be near shell 2 and the unperforated wall 15 would have a diameter smaller than the diameter of wall 8.

The present invention can be obviously exploited also when
5 a reduction in the production capacity of the existing reactor is desired, and therefore when a reduction in the mass of the (low yield) conventional catalyst to be loaded in the reactor is required.

*** * ***

10 From what has been expounded hereinabove, the many advantages achieved by the present invention become apparent; in particular, it is possible to load only partly a catalytic bed of the radial or axial-radial type of a
15 pre-existing reactor, obtaining in this way a saving in the cost of the catalyst, without affecting thereby the running of the reactor adversely.

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CLAIMS

1. Method for in-situ modernization of a heterogeneous synthesis reactor, including an external shell comprising at least a catalytic bed (6) of the radial or axial-radial type, provided with opposite perforated cylindrical walls for the inlet and outlet (7, 8) of gases, said method comprising the steps of:
- 5 - providing an unperforated cylindrical wall (15) coaxial to said gas outlet wall (8) in said catalytic bed (6), said unperforated cylindrical wall (15) extending from an upper end (8a) of said gas outlet wall (8) for a portion of the same of a prefixed length, so as to define a free-space (16) between the gas outlet wall (8) and the unperforated wall (15), for the passage of a part of the gas leaving 10 said catalytic bed (6);
- 15 - proving means of closing said free-space (16) between the unperforated wall (15) and the gas outlet wall (8), in proximity of the upper end (8a) of the latter, preventing thereby a bypass of said catalytic bed or a recycling to 20 the same of the gas entering respectively leaving the reactor.
2. Method according to claim 1, characterised in that said unperforated wall extends for a portion comprised between 5% and 50% the length of said gas outlet wall (8).
- 25 3. Method according to claim 1, characterised in that said free-space (16) has a thickness comprised between 0,5 and 10 cm.
- 30 4. Method according to claim 1, characterised in that said unperforated wall (15) is supported by said gas outlet wall (8).
5. Method according to claim 4, wherein said gas outlet wall (8) has a diameter smaller than the diameter of said

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gas inlet wall (7) and of said unperforated wall (15) respectively, characterised in that said unperforated wall (15) is supported by a gas-tight horizontal baffle (17) which protrudes above the upper end (8a) of said gas outlet wall (8), and leans on the same.

- 5
6. Heterogeneous synthesis reactor of the type comprising:
- an external shell (2);
 - at least a catalytic bed (6) of the radial or axial-radial type, provided with opposite perforated cylindrical walls for the inlet and outlet (7, 8) of gases, extended in 10 said shell (2);
- characterized in that it further comprises in said catalytic bed:
- an unperforated cylindrical wall (15) coaxial to said gas 15 outlet wall (8) in said catalytic bed (6), said unperforated cylindrical wall (15) extending from an upper end (8a) of said gas outlet wall (8) for a portion of the same of a prefixed length, so as to define a free-space (16) between the gas outlet wall (8) and the unperforated 20 wall (15), for the passage of a part of the gas leaving said catalytic bed (6);
 - means of closing said free-space (16) between the unperforated wall (15) and the gas outlet wall (8), in proximity of the upper end (8a) of the latter, preventing 25 thereby a bypass of said catalytic bed or a recycling to the same of the gas entering respectively leaving the reactor.
7. Reactor according to claim 6, characterised in that said unperforated wall (15) extends for a length comprised 30 between 5% and 50% the length of said gas outlet wall (8).
8. Reactor according to claim 6, characterised in that said

free-space (16) is substantially annular and has a thickness comprised between 0,5 and 10 cm.

9. Reactor according to claim 6, characterised in that said unperforated wall (15) is supported by said gas outlet wall 5 (8).

10. Reactor according to claim 9, wherein said gas outlet wall (8) has a diameter smaller than the diameter of said gas inlet wall (7) and of said unperforated wall (15) respectively, characterised in that said unperforated wall (15) is supported by a gas-tight horizontal baffle (17) which protrudes above the upper end (8a) of said gas outlet wall (8), and leans on the same.

ABSTRACT

A method for in-situ modernization of a heterogeneous synthesis reactor, comprises the steps of providing an unperforated cylindrical wall (15) coaxial to the gas outlet wall (8) in a catalytic bed (6) of the radial or axial-radial type, the unperforated cylindrical wall (15) extending from an upper end (8a) of the gas outlet wall (8) for a portion of the same of a prefixed length, so as to define a free-space (16) between the gas outlet wall (8) and the unperforated wall (15), for the passage of a part of the gas leaving said catalytic bed (6), and of proving means of closing the free-space (16) between the unperforated wall (15) and the gas outlet wall (8), in proximity of the upper end (8a) of the latter, preventing thereby a bypass of the catalytic bed or a recycling to the same of the gas entering respectively leaving the reactor. Thanks to the above steps, the present method allows to operate with a reduced amount of catalyst in the catalytic bed, keeping however the fluid dynamics and pressure drop characteristics unchanged.

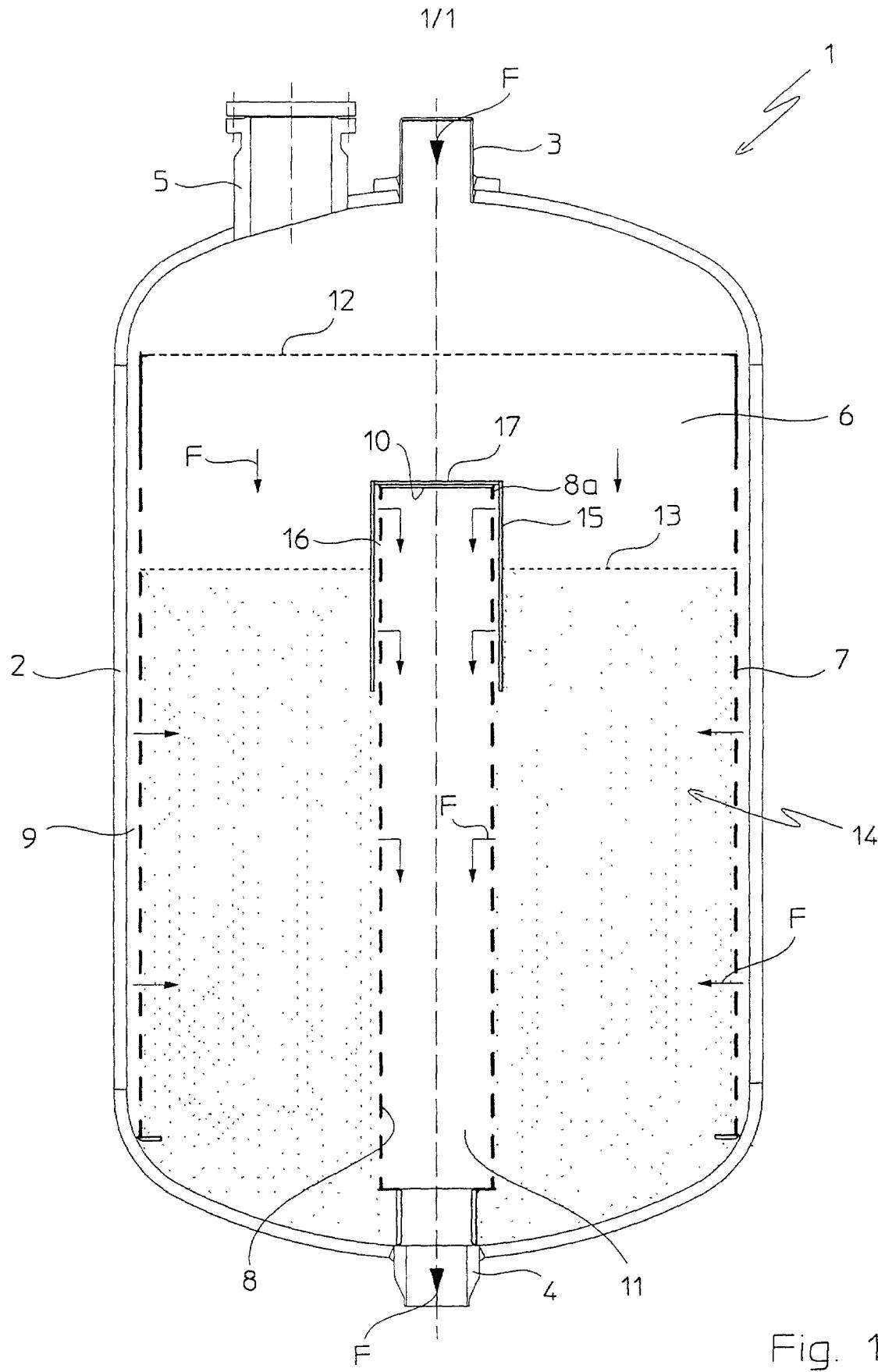


Fig. 1

Declaration and Power of Attorney for Patent Application

R-35

Dichiarazione e procura ai fini della domanda di brevetto

Italian Language Declaration

Il sottoscritto inventore dichiara che:

La propria residenza, recapito postale e cittadinanza corrispondono a quanto indicato in calce, sotto la propria firma.

Ritiene di essere il primo ed unico inventore originale (se viene elencato in calce un solo nominativo) o il coinventore primo ed originale (se è elencato più di un nominativo) del oggetto rivendicato e per il quale il sottoscritto presenta domanda di brevetto. La invenzione in questione è chiamata

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

e la sua descrizione è allegata alla presente Dichiarazione a meno che non sia spuntata la seguente casella:

- Il _____ è stata depositata una domanda di brevetto statunitense numero o una domanda di brevetto internazionale PCT numero _____ che è stata modificata il _____ (se applicabile).

the specification of which is attached hereto unless the following box is checked:

- was filed on _____ as United States Application Number or PCT International Application Number _____ and was amended on _____ (if applicable).

Il sottoscritto dichiara in oltre di aver letto e compreso il contenuto della descrizione identificata in precedenza, rivendicazioni comprese, come modificati dall'eventuale modifica summenzionata.

Il sottoscritto riconosce l'obbligo di rivelare informazioni essenziali ai fini della determinazione della brevettabilità si sensi del Titolo 37, Codice dei Regolamenti Federali, § 1.56.

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56.

[Page 1 of ____]

Burden Hour Statement: This form is estimated to take 0.4 hours to complete. Time will vary depending upon the needs of the individual case. Any comments on the amount of time you are required to complete this form should be sent to the Chief Information Officer, Patent and Trademark Office, Washington, DC 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Commissioner of Patents and Trademarks, Washington, DC 20231.

Italian Language Declaration

Il sottoscritto rivendica con la presente la priorità prevista dal Titolo 35, Codice degli Stati Uniti, § 119(e)-(d) o § 365(b) in relazione a qualsiasi domanda o domande estere di brevetto o certificato di inventore, o dal Titolo 35, § 365(a) degli stessi Codice in relazione a qualsiasi domanda internazionale PCT nella quale è designato almeno un paese diverso dagli Stati Uniti, i suddetti domande e certificati essendo elencati sotto, e, spuntando le seguenti caselle, ha anche identificato sotto qualsiasi domanda estera di brevetto o certificato di inventore, o domanda internazionale PCT, la cui data di deposito prececa quella della domanda per la quale è rivendicata la priorità.

**Prior foreign application(s)
Domande Estere Anteriori**

(Number) (Numero)	(Country) (Nazione)
98 200 114.1	Europe
(Number) (Numero)	(Country) (Nazione)

Il sottoscritto rivendica con la presente i benefici previsti dal Titolo 35, Codici degli Stati Uniti, § 119(e), in relazione a qualsiasi domanda o domande provvisorie degli Stati Uniti elencate sotto.

(Application No.) (N° della domanda)	(Filing Date) (Data di deposito)

Il sottoscritto rivendica con la presente i benefici previsti dal Titolo 35, Codice degli Stati Uniti, § 120, in relazione a qualsiasi domanda o domande statunitensi, o dal Titolo 35, § 365(c) degli stessi Codice in relazione a qualsiasi domanda internazionale PCT nella quale sono designati gli Stati Uniti, i suddette domande essendo elencate sotto e, nella misura in cui l'oggetto di ciascuna rivendicazione di questa domanda non sia stato esposto nella domanda statunitense o internazionale PCT anteriore nel modo previsto dal primo paragrafo del Titolo 35, Codice degli Stati Uniti, § 112, riconosce l'obbligo di rivelare informazioni essenziali ai fini della determinazione della brevettabilità ai sensi del Titolo 37, Codici dei Regolamenti Federali, § 1.56, le quali diventino disponibili durante il periodo compreso tra la data di deposito della domanda anteriore e la data di deposito nazionale o internazionale PCT della presente domanda.

(Application No.) (N° della domanda)	(Filing Date) (Data di deposito)

Con la presente, il sottoscritto dichiara veritiere tutte le affermazioni contenute in questa domanda in relazione alle proprie conoscenze e di ritenere vere tutte le affermazioni o informazioni presentate. Dichiara inoltre che tali asserzioni sono state espresse nella piena consapevolezza che le dichiarazioni intenzionalmente false sono punibili con una multa, l'incarcerazione o entrambe, ai sensi della Sezione 1001 del Titolo 18 del Codice degli Stati Uniti e che tali dichiarazioni intenzionalmente false possono mettere a repentaglio la validità della domanda o di qualsiasi brevetto rilasciato in merito.

I hereby claim foreign priority under Title 35, United States Code, § 119(a)-(d) or § 365 (b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

**Priority Not Claimed
Diritto di priorità non rivendicato**

<input type="checkbox"/>
(Day/Month/Year Filed) (Giorno/Mese/Anno di deposito)
16 January 1998 (Day/Month/Year Filed) (Giorno/Mese/Anno di deposito)

I hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States provisional application(s) listed below.

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

**(Status)(patented, pending, abandoned)
(Stato)(concessione di brevetto, in corso di esame, abbandono)**

**(Status)(patented, pending, abandoned)
(Stato)(concessione di brevetto, in corso di esame, abbandono)**

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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Italian Language Declaration

PROCURA: Il sottoscritto inventore nomina con la presente il seguente avvocato o avvocati e/o agente o agenti al fine di istruire questa pratica e di condurre tutte le operazioni ad essa pertinenti presso l'Ufficio dei Brevetti e Marchi di Fabbrica: (*Elencare il nome ed il numero di matricola*)

John H. Mion, Reg. No. 18,879; Donald E. Zina, Reg. No. 19,046; Thomas J. Macpeak, Reg. No. 19,292; Robert J. Sosa, Jr., Reg. No. 21,092; Darryl Mezie, Reg. No. 23,063; Robert V. Slous, Reg. No. 22,775; Peter D. Olcay, Reg. No. 24,513; J. Frank Goba, Reg. No. 24,625; Weddell A. Biggart, Reg. No. 24,881; Robert O. McCormick, Reg. No. 19,095; Louis Cukinsky, Reg. No. 24,835; Neil B. Siegel, Reg. No. 25,200; David J. Cushing, Reg. No. 28,703; John R. Inga, Reg. No. 26,916; Joseph J. Ruch, Jr., Reg. No. 26,577; Sheldon I. Landman, Reg. No. 25,430; Richard C. Turner, Reg. No. 29,710; Howard L. Bernstein, Reg. No. 25,665; Alan J. Kasper, Reg. No. 25,426; Kenneth J. Burchfield, Reg. No. 31,333; Gordon K. Reg. No. 30,764; Susan J. Mack, Reg. No. 30,951; Frank L. Bernstein, Reg. No. 31,484; Mark Boland, Reg. No. 32,197; William H. Mandel, Reg. No. 32,156; Scott M. Daniels, Reg. No. 32,562; Bruce W. Hawson, Reg. No. 32,778; Abraham J. Rosner, Reg. No. 33,276; Bruce E. Kramer, Reg. No. 33,725; Paul F. Neils, Reg. No. 33,102; and Brett S. Sylvester, Reg. No. 32,765.

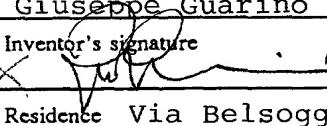
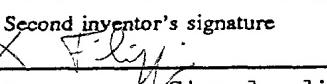
POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith: (*list name and registration number*)

Inviare le corrispondenza a:

Send Correspondence to:

SUGHRUE, MION, ZINN, MACPEAK & SEAS, PLLC
2100 Pennsylvania Avenue, N.W. Suite 800
Washington, D.C. 20037-3202

Telefonare a: (*nome e numero telefonico*)Direct Telephone Calls to: (*name and telephone number*)

Nome e cognome dell'unico o del primo inventore	Full name of sole or first inventor Giuseppe Guarino	
Firma dell'inventore	Data	Inventor's signature  Date 15.12.1998
Residenza	Residence Via Belsoggiorno 6965 Cadro, Switzerland	
Cittadinanza	Citizenship Italian	
Recapito postale	Post Office Address	
Nome e cognome dell'eventuale secondo coinventore	Full name of second joint inventor, if any Ermanno Filippi	
Firma del secondo coinventore	Data	Second inventor's signature  Date 15.12.1998
Residenza	Residence Strada di Gandria 73 6976 Castagnola, Switzerland	
Cittadinanza	Citizenship Italian	
Recapito postale	Post Office Address	

(Fornire le stesse informazioni e le firme del terzo e degli ulteriori coinventori.)

(Supply similar information and signature for third and subsequent joint inventors.)

Applicant or Patentee: Giuseppe GUARINO et al.
 Serial or Patent No.: _____

Attorney's Docket No.: Q-52856

Filed or Issued:

Title: Method for in-situ modernization of a heterogeneous synthesis reactor

VERIFIED STATEMENT (DECLARATION) CLAIMING SMALL ENTITY STATUS
 (37 CFR 1.9(f) & 1.27(c))--SMALL BUSINESS CONCERN

I hereby declare that I am

- the owner of the small business concern identified below:
 an official of the small business concern empowered to act on behalf of the concern identified below:

NAME OF SMALL BUSINESS CONCERN Ammonia Casale SA
 ADDRESS OF SMALL BUSINESS CONCERN Via Sorengo 7, 6900 Lugano-Besso
 Switzerland

I hereby declare that the above identified small business concern qualifies as a small business concern as defined in 13 CFR 121.12, and reproduced in 37 CFR 1.9(d), for purposes of paying reduced fees to the United States Patent and Trademark Office, in that the number of employees of the concern, including those of its affiliates, does not exceed 500 persons. For purposes of this statement, (1) the number of employees of the business concern is the average over the previous fiscal year of the concern of the persons employed on a full-time, part-time or temporary basis during each of the pay periods of the fiscal year, and (2) concerns are affiliates of each other when either, directly or indirectly, one concern controls or has the power to control the other, or a third party or parties controls or has the power to control both.

I hereby declare that rights under contract or law have been conveyed to and remain with the small business concern identified above with regard to the invention, entitled Method for in-situ modernization of a heterogeneous synthesis reactor invented by Giuseppe Guarino, Ermanno Filippi, described in

- the specification filed herewith
 application serial no. _____ filed _____
 patent no. _____ issued _____

If the rights held by the above identified small business concern are not exclusive, each individual, concern or organization having rights in the invention is listed below* and no rights to the invention are held by any person, other than the inventor, who would not qualify as an independent inventor under 37 CFR 1.9(c) if that person made the invention, or by any concern which would not qualify as a small business concern under 37 CFR 1.9(d), or a nonprofit organization under 37 CFR 1.9(e). *NOTE: Separate verified statements are required from each named person, concern or organization having rights to the invention averring to their status as small entities. (37 CFR 1.27)

NAME _____
 ADDRESS _____
 INDIVIDUAL SMALL BUSINESS CONCERN NONPROFIT ORGANIZATION

NAME _____
 ADDRESS _____
 INDIVIDUAL SMALL BUSINESS CONCERN NONPROFIT ORGANIZATION

I acknowledge the duty to file, in this application or patent, notification of any change in status resulting in loss of entitlement to small entity status prior to paying, or at the time of paying, the earliest of the issue fee or any maintenance fee due after the date on which status as a small entity is no longer appropriate. (37 CFR 1.28(b))

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application, any patent issuing thereon, or any patent to which this verified statement is directed.

NAME OF PERSON SIGNING Umberto Zardi
 TITLE OF PERSON IF OTHER THAN OWNER Managing Director
 ADDRESS OF PERSON SIGNING _____

SIGNATURE X Mary DATE 15.12.1998